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ANGELES DEPT OF MATERIALS SCIENCE AND ENGINEERING
F F LANGE ET AL. 13 MAY 85 N00014-84-K-0286

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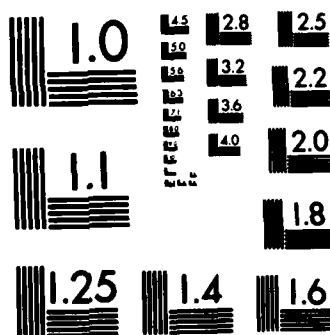
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SINTERING STUDIES PROJECT

Sponsored by ONR N00014-84-K-0286

Research Progress during 1 April 1984 - 31 March 1985

Principal Investigators:

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I. INTRODUCTION

Previous work⁽¹⁾ introduced a new concept to explain the factors which influence the sinterability of ceramic (and metal) powders. This new theory, based on the thermodynamics consideration of pore disappearance, is centered on the coordination number of distribution of pores within the initial powder compact and on the changes in the pore coordination number during sintering. Thermodynamic arguments show that only pores with a coordination number less than a critical value ($R > R_c$) can disappear by diffusion.

The new concept suggested the following phenomena:

- (1) Consolidation forces (e.g., iso-pressing) that increase bulk density will eliminate pores with high coordination numbers and thus increase the sinterability of the powder compact.
- (2) Multiple-particle packing units containing pores with low coordination ($R < R_c$) will first densify during sintering to increase the size and coordination number of pores between the densifying packing units.
- (3) Grain growth, supported by dense packing units, will decrease the coordination number of pores enlarged by local densification. ~~Namely~~ i.e. normal grain growth is advantageous to sinterability.
- (4) Compressive stress produced by either surface tension effects or hot-pressing cause the dense regions to deform, thus closing pores between the dense regions and reducing their coordination number.

(*The coordination number of a pore is simply the number of touching particles that surround and the pore size.)

**Abnormal grain growth which entrap pores is undesirable

Work done in the first year of this contract focussed on several areas: (i) theoretical modeling of the shrinkage of particle clusters, (ii) experimental work concerned with influence of grain growth on pore disappearance during sintering, and (iii) direct observation of sintering. The following will outline the current results of these areas.

II. THEORETICAL MODELING

As is well known, powder compacts are not perfectly packed, and because of differences in packing, even within the same compact, complexities arise; for instance, nonuniform sintering within a powder compact can cause the formation of large pores. Mercury porosimetry has shown that the average pore size usually increases with sintering. This behavior can not be explained by assuming uniform packing and sintering within the powder compact.

To consider the effect on densification, we have viewed sintering as the interpenetration of spheres. The contact angle formed between two neighboring spheres, at the grain boundary, increases as the spheres interpenetrate; this contact angle increases, from 0° in the green state, to the dihedral angle ϕ at equilibrium. We have considered idealized models, where monosized spheres have been placed in various configurations. When these spheres are arranged in a ring configuration, the pore region within the ring is found to become unstable at critical contact angle. The critical angle increases with the number of surrounding spheres (coordination).

This simple model indicates how the geometric natures of particle packing within the powder compact affects densification, and suggests that

sintering cannot be described by mass transport equations alone. If sintering was solely diffusion-controlled, then pores of all sizes would all shrink at some constant rate. By our calculations, large pores with coordination numbers greater than $2\pi/(\pi-\phi)$ would shrink to an equilibrium size, dictated by the coordination number and the dihedral angle, (ϕ) , and would remain at this size until their coordination, lowered by grain growth, reaches $2\pi/(\pi-\phi)$. The pore at this point becomes unstable and is eliminated by mass transport.

Pore stability was determined by considering closed arrays of identical particles and calculating their equilibrium configuration. These calculations indicated that

- a. all pores shrink from their initial configuration
- b. only pores which have less than a critical coordination are thermodynamically unstable and disappear.

The critical coordination for a ring of particles is $n = 2\pi/(\pi-\phi)$, where $2\cos\frac{\phi}{2} = \sigma_{ss}/\sigma_{sv}$, and σ_{ss} and σ_{sv} are grain boundary and surface energies, respectively.

III GRAIN GROWTH AND PORE DISAPPEARANCE

Polystyrene spheres with diameters of 1, 2 and 4 μ m were used to introduce spherical pores into an Al_2O_3/ZrO_2 material. (The ZrO_2 second phase (10 vol %) serves to slow down and control the grain growth of the material.) After sintering at 1600C for 5 min, only the purposely introduced spherical pores remained. Specimens with different pore diameters were heat treated at 1600°C

to 1700°C to induce grain growth. The grain and pore size distributions, after polishing and thermal etching, were measured. Heat treatment increased the average grain size and skewed the distribution at larger grain sizes. The mean size of the purposely introduced pores remained constant throughout the heat treatment, but the number of pores per unit area was dramatically decreased. These results are consistent with thermodynamic argument that pores will only disappear when they became coordinated by a critical number of grains.

Work performed more recently has expanded on these ideas by considering the effect of pressure, uniaxial stress, and surface energy anisotropy on pore stability.

Uniaxial stress were applied to specimens containing model pores by use of a high temperature creep furnace. Preliminary results indicate that pores are closed only by deformation of the order of $\gamma = 0.7$. To get strains of this magnitude requires that the ceramic exhibit slow and controlled grain growth. Creep of polycrystalline solids is strongly a function of grain size, with greater strain rates obtainable with finer grain material. Consequently sufficient creep strains were obtained for two phase $\text{Al}_2\text{O}_3/\text{ZrO}_2$ systems, and were not obtained for single phase Al_2O_3 . The pore shape, observed on polished and etched surfaces, is initially circular in cross section, and with applied strain becomes flattened, finally becoming a flat dish-shape in which grains from the top surface come into contact with the bottom. At strains of about 0.7, all that is left of the pore are clusters of smaller pores, and these disappear with reactivity little extra strain. The pore, while shrinking along the stress axis, was found to maintain its width. The

development of this geometry is similar to that calculated from a model which assumes that the material is perfectly Newtonian in nature.

IV OBSERVATION OF THE SINTERING OF MICROSPHERES

Independent experiments have been initiated, which enable us to observe directly the sintering of microspheres, for correlation with models. Uniform microspheres of glass, packed in a single layer on the hot stage (Leitz #1350) of an optical microscope, have been observed during heating. The progress of sintering is being recorded by photographs, and, if advantageous, by videotape. Glass microspheres of several diameters (about 43 μ m and 110 μ m), were packed together, in an attempt to produce several pore geometries, which were observed during sintering. This experiment is similar, in a general way, to prior studies of Exner⁽²⁾ and Sacks and Tseng,⁽³⁾ excepting that the proposed experiment enables continuous observation (and photography) during the progress of sintering. An alternative study material is offered by microspheres of polystyrene, which Lange has found will sinter at a temperature slightly above 200°C; the sintering of such microspheres is in progress.



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V PRESENTATIONS AND PAPERS RESULTING FROM THIS RESEARCH (During the contract period 1 April 1984 - 31 March 1985)

Presentations

1. "Model Pores," B. J. Kellett* and F. F. Lange, 17th Pacific Coast Regional Meeting, American Ceramic Society, San Francisco, CA, 29 October 1984.
2. "Influence of Particle Arrangement on Sintering," F. F. Lange* and B. J. Kellett, Second International Conference on Ultrastructure Processing of Ceramic, Glasses and Composites, Palm Coast, FL, Feb. 25 - Mar. 1, 1985.
3. "Conditions for Pore Disappearance," B. J. Kellett* and F. F. Lange, Annual Meeting, American Ceramic Society, Cincinnati, OH, 8 May 1985.

Paper in Press

"Influence of Particle Arrangement on Sintering," F. F. Lange and B. J. Kellett, for the Proceedings of the Second International Conference on Ultrastructure Processing of Ceramics, Glasses and Composites.

Papers in Preparation

1. "Model Pores, I. Theory," B. J. Kellett and F. F. Lange (for American Ceramic Society).
2. "Model Pores, II. Experiment," B. J. Kellett and F. F. Lange (for American Ceramics Society).

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1. F. F. Lange, "Sinterability of Agglomerated Powders," Materials Research Society Symposium Proceedings, Vol. 24 (1984) Elsevier.
2. H. E. Exner, "Principles of Single Phase Sintering," Reviews on Powder Metallurgy and Physical Ceramics, 1[1-4], 1-251 (1979).
3. M. D. Sacks and T-Y Tseng, "Preparation of SiO_2 Glass from Model Powder Compacts: II, Sintering," J. American Ceramic Soc., 67[8] 532-7 (1984).

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